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Enhancement of Long Range Antiferromagnetic Order by Nonmagnetic Impurities in the Hubbard Model

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Abstract. –

The two–dimensional Hubbard model with a bimodal distribution of on–site interactions, $P(U_{\mathbf{i}}) = (1-f)\delta(U_{\mathbf{i}} - U) + f\delta(U_{\mathbf{i}})$, is studied using finite temperature Quantum Monte Carlo and dynamical mean–field theory. Long range antiferromagnetic order off half–filling is *stabilized* by the disorder, due to localization of the dopants on the $U = 0$ sites. Whereas in the clean model there is a single gap at $n = 1$, for nonzero f we find the compressibility and density of states exhibit gaps at two separate fillings.

The Hubbard model exhibits both Mott metal–insulator and magnetic phase transitions. On the one hand, the interaction U induces a gap at half–filling by separating many–body states with doubly occupied orbitals (upper Hubbard band) from those with holes (lower Hubbard band) when U becomes larger than the non–interacting bandwidth W. On the other hand, also near half–filling, there is a tendency to antiferromagnetic (AF) ordering of the electron spins. For interacting fermions, the issue has recently been raised whether the Mott transition ever occurs in the absence of associated symmetry breaking such as magnetic order [1].

The introduction of "impurity" sites where $U = 0$ in principle could separate AF order and the Mott transition. The Mott gap will be shifted to densities greater than $n = 1$ since some sites can be doubly occupied without any on–site repulsion energy cost. Meanwhile, it is likely that the Fermi–surface instability responsible for opening the AF (spin density wave) gap remains at half–filling. In this paper we will study a model Hamiltonian incorporating this effect. Our main conclusions are: (1) $U = 0$ sites can induce long range AF order at densities which are spin disordered in the clean model, through localization of the doped particles. (2) The dependence of the occupation on the chemical potential and the behavior of the density of states exhibit a Mott gap, shifted from half–filling, and also a gap at half–filling resulting from induced AF order on the $U = 0$ sites.

There are a number of experimental systems where the effect of the introduction of nonmagnetic impurities has been studied. Examples include doping Zn or Ga for La in La_2CuO_4, where

the critical concentration for the destruction of AF order, $x_c \approx 0.10-0.15$ is considerably larger than for doping with Sr, which is not isovalent [2], and also Zn doping in ladder compounds where an AF phase is stabilized [3]. Numerical work on the effect of impurities on Néel order has focused on spin systems [4]. Enhanced local correlations arise from restrictions on the singlet bond patterns due to the defects [5].

We study the electronic lattice Hamiltonian,

$$\hat{H} \;=\; -t \sum_{<\mathbf{ij}>,\sigma} \hat{c}^{\dagger}_{\mathbf{i}\sigma} \hat{c}_{\mathbf{j}\sigma} - \mu \sum_{\mathbf{i}\sigma} \hat{n}_{\mathbf{i}\sigma} + \sum_{\mathbf{i}} U_{\mathbf{i}} (\hat{n}_{\mathbf{i}\uparrow} - \frac{1}{2})(\hat{n}_{\mathbf{i}\downarrow} - \frac{1}{2}) \; . \tag{1}$$

The kinetic energy is described by hopping between nearest–neighbor lattice sites, $\langle \mathbf{ij} \rangle$, on a square ($N{=}L \times L$) lattice. The interaction values $U_{\mathbf{i}}$ are chosen from a bimodal distribution, $U_{\mathbf{i}} \in \{0, U\}$, with probabilities f and $1-f$, respectively. Upon electron doping the $U = 0$ sites will be filled thereby quenching their local moment and eventually acting as magnetically inert impurities.

We use two complementary numerical approaches, finite dimensional "determinant" quantum Monte Carlo (QMC) algorithm [6, 7] and the closely related "dynamical mean field theory" (DMFT) [8, 9, 10], also employing QMC [11]. The former technique allows an approximation free solution of the equilibrium thermodynamic properties, but only on finite lattices and in a somewhat restricted range of parameters. The latter technique introduces a local approximation to the self–energy, which becomes exact in the limit of infinite dimensions, and allows a more complete exploration of parameter space in the thermodynamic limit. DMFT has proven to give reliable results for thermodynamic and dynamic properties in $d = 2$ and $d = 3$ [9, 10], and has also been applied to study disorder effects on interacting systems [12, 13]. While for $d = 2$ expectation values are averaged over typically tens of (static) disorder configurations, in DMFT this average reduces to a weighted sum over the two possible values of U_i, equivalent to the coherent–potential approximation [14].

Like the original Hubbard model, Hamiltonian (1) is particle–hole symmetric at half–filling ($n = 1$ and $\mu = 0$), i.e. it is invariant under the "staggered" particle–hole transformation $\hat{c}^{\dagger}_{\mathbf{i}\sigma} \rightarrow (-1)^{\mathbf{i}} \hat{c}_{\mathbf{i}\sigma}$, so there is no minus–sign problem at $n = 1$ which would preclude simulations for large lattices at low temperatures T. Physically, the particle–hole symmetry corresponds to different chemical potentials on the two constituents such that at $\mu = 0$ the local density expectation values $n_{\mathbf{i}}$ are homogeneous, i.e. independent of $U_{\mathbf{i}}$ [15]. The choice here of the symmetric limit of the model is identical in spirit to the focus in the heavy fermion literature on the 'symmetric' Anderson model, in which a similar specific relation between f-site energy and f interaction strength is assumed to bring out most clearly the physics of that Hamiltonian. In the following we will discuss the effects of disorder on (i) AF order and (ii) the charge gap, presenting for each case first $d = 2$ data which are then complemented by DMFT results.

AF order. — First we consider the case $n = 1$ and study the effect of an increasing concentration f of $U = 0$ sites on the stability of AF long range order. The static AF structure factor $S(\pi, \pi)$ is calculated for different lattice sizes ($L \leq 10$) and temperatures, and averaged over up to 40 disorder configurations. For $L \leq 10$, $S(\pi, \pi)$ is found to saturate at $T \approx t/8$. From the saturated values the ground state sublattice magnetization M can be extrapolated using a finite–size scaling according to spin wave theory [16], $S(\pi, \pi)/L^2 = M^2/3 + O(1/L)$. Scaling plots for different f at $U = 8t$ are shown in the inset of Fig. 1. For $f \leq 0.36$, $S(\pi, \pi)/L^2$ extrapolates to a finite value in the thermodynamic limit. For $f = 0.5$ the linear scaling with $1/L$ does not hold anymore, since it leads to negative values of M^2, indicating the absence of AF order in the thermodynamic limit. Fig. 1 presents the extrapolated values of M as a

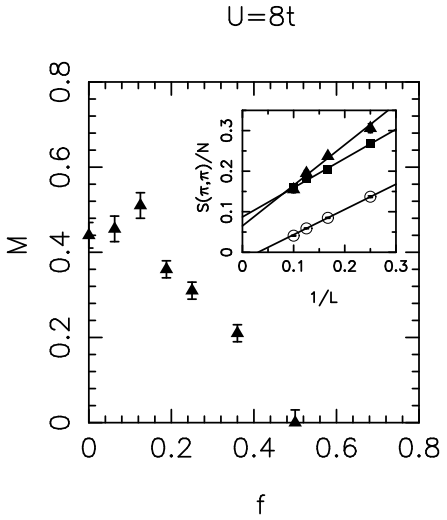
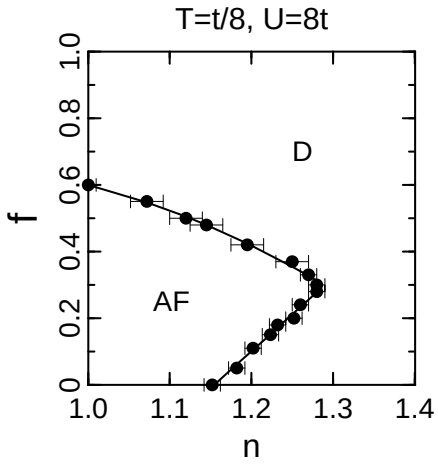


Fig. 1. – Ground state staggered magnetization M as a function of fraction of $U = 0$ sites, f, for $U = 8t$, $n = 1$. Inset: Finite–size scaling of the antiferromagnetic structure factor. f =0.0, 0.125, (filled triangles and squares) are ordered. $f = 0.500$ (open circles) is not. For values of f that do not correspond to an integer number of defects, we have interpolated the results for the two bracketing concentrations.

function of f. A spin wave suppression of the staggered magnetization[19]. might be the origin of the weak enhancement of M near $f = 0.1$.

The $U = 0$ impurity sites can induce AF order at a doping for which the clean model is spin disordered. At finite doping the minus–sign problem prevents the simulations necessary for the finite–size analysis. We therefore employ DMFT to calculate the phase diagram in the density–disorder plane, shown in Fig. 2 for $T = t/8$. The phase boundary is obtained both from the vanishing of M in the AF phase and from the divergence of the staggered susceptibility in the spin disordered (D) phase. The region of AF long range order extends to a larger doping for finite f; the presence of impurity sites stabilizes order. In the clean model, doping away from half–filling introduces extra particles which are mobile and hence especially effective at disturbing correlations over the entire lattice. $U = 0$ defects provide localizing sites which are energetically favorable for these extra particles. Thus, while doping destroys moments locally, it has a much smaller effect on long range correlations. In two dimensions, the critical doping value for the clean $f = 0$ model is smaller than in DMFT and probably zero because of competing charge or incommensurate magnetic instabilities. However, we expect that the enhancement of the AF phase by nonzero f will also be present in $d = 2$, since the underlying mechanism, the defect–induced localization, is observed in $d = 2$, too.

Charge gap. — In the extreme strong coupling limit, $t = 0$, a plot of the density n as a function of chemical potential μ exhibits plateaus at $n = 1 \pm f$: n is zero until $\mu = -U/2$, at which point n will jump to $n = 1 - f$ (singly occupying sites which have nonzero U.) When μ goes through zero, n will jump to $n = 1 - f + 2f = 1 + f$ (singly occupied nonzero U sites plus doubly occupied $U = 0$ sites). Finally, at $\mu = +U/2$, the density jumps to $n = 2$ (all sites doubly occupied). When t is turned on these sharp steps will be rounded by quantum fluctuations, but we might expect the compressibility $\kappa = \partial n/\partial\mu$ to remain large at $n = 1$ and remain small at $n = 1 + f$. In order to explore precisely the effect of nonzero hopping the dependence of n on the chemical potential is calculated using $d = 2$ QMC. Fig. 3 shows results



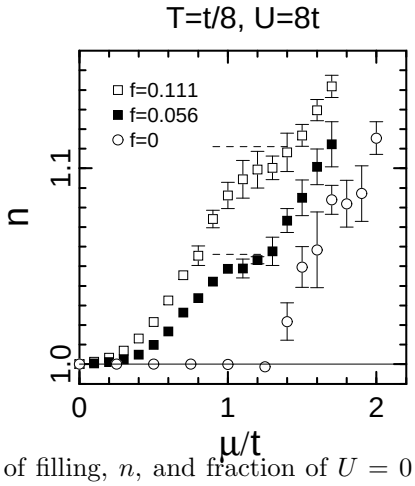


Fig. 2. – Phase diagram in DMFT as a function of filling, n, and fraction of $U = 0$ sites, f. For $0 < f < 0.4$ the antiferromagnetic (AF) region around half–filling is enhanced relative to $f = 0$. At fixed density $n > 1$, the introduction of $U = 0$ impurities localizes the mobile dopants and restores long range order.

Fig. 3. – Density n vs. chemical potential μ for different values of f on a 6×6 lattice with $U = 8t$. Error bars are within the symbol size when not shown [18]. The Mott plateau is shifted away from half–filling. Dashed lines indicate the values $1 + f$ (corresponding to complete occupation of $U = 0$ sites).

for a $6 \times 6-$lattice at $T = t/8$ and $U = 8t$ [17]. At $f = 0$ there is a wide charge gap, visible in the broad plateau at $n = 1$ starting at $\mu = 0$. For $f > 0$ the "Mott" gap occurs close to the expected density of $n = 1 + f$ (dashed lines). It terminates at a chemical potential close to the μ_c for $f = 0$. That is, the chemical potential to force double occupation of the non–zero U sites is unaffected by the presence of the $U = 0$ sites. In addition, however, a second plateau remains at half–filling, an effect not predicted by the $t = 0$ analysis.

In order to elucidate the nature of this remnant gap at half–filling, we examine the effect of the chemical potential on the staggered magnetization M. Fig. 4 shows DMFT results for the densities and sublattice magnetizations vs. μ at $f = 0.11$ for $U = 0$ and $U = 8t$ sites separately. The densities show quantitatively the same dependence on μ in $d = 2$ and in DMFT [18], however the sublattice magnetization off half–filling can be obtained in DMFT only. The density on the $U = 8t$ sites hardly changes for $\mu < 1.6t$, but then begins to rise, at which point M also abruptly vanishes. For small μ there is induced AF order also on the $U = 0$ sites. This AF order leads to a gap even on the $U = 0$ sites due to the doubling of the unit cell. The two different gap values on the $U = 8t$ and $U = 0$ sites explain the double gap structure in the total density (Fig. 3).

Surprisingly, M becomes *negative* on the $U = 0$ sites when $n(U = 0)$ starts to saturate. The reason is that an electron on a $U = 0$ site with spin parallel to its neighbors is more strongly localized (due to Pauli's principle) than an electron with opposite spin. Thus the net moment on the $U = 0$ site is *parallel* to its neighbors, i.e. opposite to the total staggered magnetization. Within the DMFT approach AF order is more stable against disorder ($f_c^\infty = 0.75 \pm 0.02$ at $T = 0$ and $n = 1$, compared to $f_c \approx 0.4$ in $d = 2$), as is typical for mean–field theories.

The charge gap can also be obtained from the density of states (DOS). We employ the

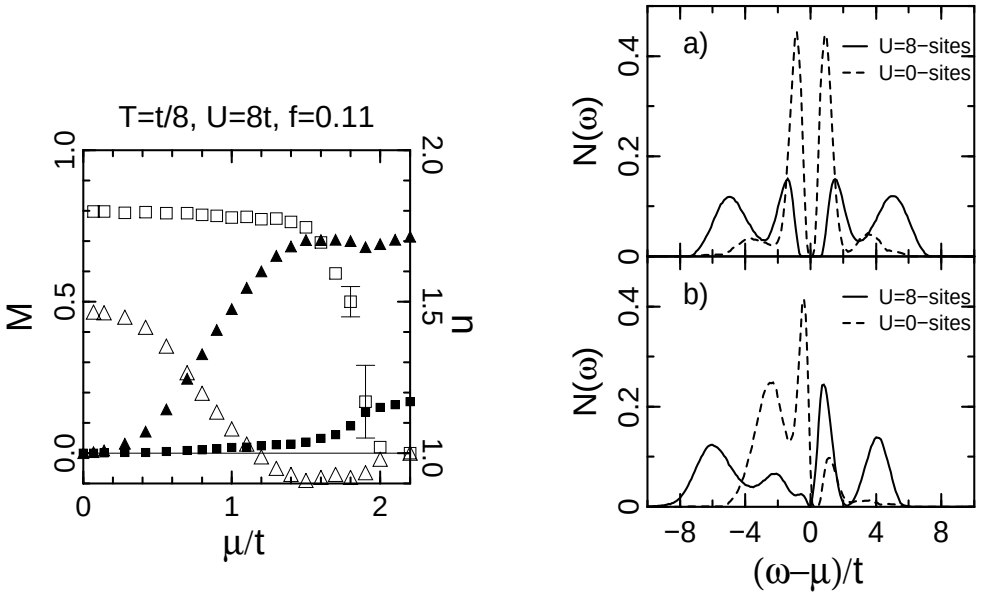


Fig. 4. – Density n (solid symbols) and sublattice magnetization M (open symbols) vs. μ in DMFT separately for $U = 0$ sites (triangles) and $U = 8t$ sites (squares). For small μ there is AF order on both types of sites. For $\mu > 1.6t$ the density on the $U = 8t$ sites increases and AF order breaks down.

Fig. 5. – Density of states in DMFT on $U = 0$ and $U = 8t$ sites for $T = t/8$, $f = 0.11$ at a) half–filling and b) $n = 1.11$. All spectra have a vanishing DOS at the Fermi energy.

DMFT approach, combined with the "Maximum Entropy" method [22], which gives excellent agreement with $d = 2$ and $d = 3$ results in the clean case [20, 21]. At half–filling and low T (Fig. 5a) the DOS for the $U = 8t$ sites shows the typical AF structure [21] with two broad Hubbard bands, two quasi–particle peaks at low energies, and a gap at the Fermi energy, $E_F = 0$. On the $U = 0$ sites the Hubbard bands appear at lower energies and the gap is very small but still present, and will increase at lower T. Thus there is indeed a gap in the antiferromagnetic phase, even for the $U = 0$ sites. For small doping ($1.0 < n < 1.11$) the compressibility was seen to be large (see Figs. 3 and 4). At $n = 1.11$, where the density on the $U = 0$ sites saturates, the DOS again vanishes at the Fermi energy, $E_F = \mu = 1.45t$ (Fig. 5b). At this point there is still AF order.

Summary. — We can summarize the physics of the charge gap and the AF order in terms of the phase diagram of Fig. 2. In the pure case AF order breaks down when the density reaches a critical value $n_c(0)$. This value depends on dimensionality and may very well be 1.0 in $d = 2$. At a finite concentration f, the defect sites localize dopants, thereby keeping the rest of lattice at half filling, until they are (almost) doubly occupied. For small concentration f the defect sites hence stabilize AF order up to a total density of $n_c(f) \approx n_c(0)(1 - f) + 2f$. Eventually, at large enough f, n_c is driven to one and AF order ceases to exist even for $n = 1$. The density exhibits a plateau associated with AF order, which exists on both the nonzero U and the $U = 0$ sites, as the chemical potential is changed across half–filling ($n = 1$); and a second "Mott" plateau, associated with double occupancy, as one crosses the high filling

boundary of the AF region.

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